Vapor Deposition and Etching Under Diffusion-Controlled Process Conditions

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Summary

The objective of the envisioned experiment in space is to validate, on the basis of results of optical real-time monitoring process models, simulation codes, and input parameter, sets that are needed for analysis and control of chemical vapor deposition and etching at elevated pressure. Access to microgravity is required to retain high pressure conditions of laminar flow, which is essential for successful acquisition and interpretation of the optical data. The extension of chemical vapor deposition or etching to elevated pressure is necessary for retaining stoichiometric single phase surface composition for materials that are characterized by large thermal decomposition pressures at optimum processing temperatures.

We target InN as model material, which, under conditions of conventional organometallic chemical vapor deposition (OMCVD) at subatmospheric pressure, is limited to processing temperatures 800 K. This restriction, in turn, limits the control of extended defects in this material and is one of the reasons why, at present, confined $Ga_xIn_{1-x}N/AI_yGa_{1-y}N$ heterostructure devices (e.g., diode lasers) do not include In-rich compositions that maximize quantum well depth. Here we focus on deposition temperatures 900 K, which we consider as being essential for improving film quality and for improving the match between optimum deposition temperatures for quantum well and barrier layers of $Ga_xIn_{1-x}N/AI_yGa_{1-y}N$ multiple quantum well heterostructures. The desired range of processing temperatures mandates operating pressures of ~ 100 atm and above.

Based on simulations of the flow dynamics for selected high pressure OMCVD reactor designs, we present the design of an optimized channel flow reactor for the envisioned microgravity experiment. Also, we present a ground-based OMCVD system that has been constructed in the PI's laboratory (limited to operation at 7 atm pressure) and allows preliminary evaluations at low pressure. In order to avoid homogeneous nucleation of InN in the dense hot vapor atmosphere, and to permit real-time studies of the kinetics of chemical reactions, both in the gas phase and on the surface of the substrate/epilayer stack, pulsed exposure of the surface of the evolving heterostructure to plugs of source vapors separated by plugs of high pressure nitrogen carrier gas is essential. Intermixing of these pulses in regions of residual buoyancy driven recirculating flow is prohibitive and imposes a restriction on the Richardson number. This, in turn, mandates a lower limit for flow velocity and Reynolds number *Re* under ground-based conditions. Unfortunately, at p 100 atm, *Re* is large enough to cause concerns regarding onset of turbulence.

Only access to reduced gravity allows for sufficient reduction of the minimum flow velocity that is required for prevention of contamination of the flow by residual buoyancy driven recirculation (i.e., allows operation at significantly reduced Re). For the envisioned range of vapor pressures this is expected to result in laminar flow, for which adequate process models, simulation codes, and input parameters exist or can be generated with existing methods. To make sure, provisions are made in the design of the high pressure prototype reactor for implementation of experimental methods for *in-situ* characterization of the nature of flow. Also, we discuss choices of optical methods of real-time process monitoring and present results of evaluations of initial stages of heteroepitaxy and

steady-state growth at low pressure, which define upper limits for the required experiment time. Based on these results, two groups of experiments can be identified:

- 1. Experiments regarding the evaluation of initial stages of heteroepitaxy and final stages of vapor etching, requiring 10⁻³ to 10⁻⁴ g and times between 30 seconds and several minutes depending on lift-off conditions.
- 2. Experiments that address steady state growth and etching and require at least 10⁻⁵ g and several hours uninterrupted run time.

Both types of experiments are regarded as equally important and do not depend in execution and interpretation on each other.

Tasks that have highest priority in the ongoing ground-based preparations are simulations of flow in the prototype reactor at various lengths and orientations of the gravity vector, preliminary experimental studies of flow at selected values of Re in a mock-up reactor on the ground, and the calculation of essential input parameters. The latter is supported by quantum chemical calculations provided by Prof. Beatriz Cardelino of Spelman College and Dr. Craig Moore of the NASA Marshall Space Flight Center. Further support in process simulations is provided by Prof. Carlos Cardelino of the Georgia Institute of Technology.